ULTRASENSITIVE HIGH RESOLUTION LASER SPECTROSCOPY AND ITS APPLICATION TO OPTICAL FREQUENCY STANDARDS

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Abstract

Advanced laser stabilization techniques now permit one to lock laser frequencies onto line centers of natural atomic/molecular resonances with unprecedented precision and accuracy, leading to improved visible optical frequency standards. A novel approach of cavity-enhanced frequency modulation spectroscopy is demonstrated to enjoy a record high sensitivity (6×10^{-13} integrated absorption), working with a rich spectrum of weak molecular overtone transitions. The resultant high signal-to-noise ratio allows a Nd:YAG laser to be stabilized on the overtone transition at the level of 1×10^{-14} in 800 seconds. For the work of coherent optical frequency connection, we present our recent development of an optical frequency comb generator with a 3-terahertz span.

INTRODUCTION

Establishing frequency standards in the optical domain has been an extremely active field since the invention of the laser. Ultimate invariance tests of many fundamental physical postulates and constants can be approached only with the highest available optical spectral resolution, precision, and sensitivity offered by the frequency-based metrology. In addition, optical frequency references provide a direct link between the units of length and time, and they are invaluable in coherence optical communications. Owing to its higher operating frequency, it is natural to expect an optical resonance quality factor Q to be a few orders higher than those in the microwave domain. Therefore, optical frequency standards can potentially be more stable and precise. Of great long-term interest is the prospect of being able to bring into the microwave domain the superior stability and accuracy of suitable optical frequency sources.

To effectively use a laser as a stable and accurate optical local oscillator, active control of its frequency is needed due to the strong coupling between the laser frequency and its cavity. The laser's fast linewidth needs to be reduced to produce not only the desired short-term stability, but also a long coherence time appropriate for interrogation of narrow atomic or molecular transitions. Short-term stability is usually achieved by stabilizing the optical phase/frequency relative to a passive reference, such as a high-finesse stable Fabry-Perot cavity. As a fast

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frequency discriminator, the passive reference cavity is preferred over the preselected atomic or molecular resonance because of its linear response to the field and a high signal-to-noise ratio (S/N). Frequency stability at the 10^{-16} level has been measured with a cavity-stabilized laser.[1] Tunability of such a cavity/laser system can be fulfilled by techniques such as the frequency-offset optical phase-locked-loop (PLL).[2]

Long-term stability and reproducibility of the local oscillator (laser) can be obtained by locking to preselected natural resonances. Special optical techniques were invented to eliminate common sources of line broadening such as the Doppler effect. In the high resolution domain of laser spectroscopy, many research developments provide a number of sophisticated methods, such as saturated absorption spectroscopy, two-photon spectroscopy, optical Ramsey fringes, optical double resonance, quantum beat, and laser cooling and trapping.[3] Usually one or several atomic or molecular transitions are located within the tuning range of the laser to be stabilized. Molecular ro-vibrational lines are used effectively for laser stabilization in the infrared, in systems such as the methane-stabilized HeNe at 3.39 μm, and osmium-tetroxide-stabilized CO_2 lasers at 10 μm. They now typically offer a precision of 10^{-12}.[4] Until recently the molecular iodine transitions have been used exclusively as the frequency references in the visible spectrum and the precision has been limited to 10^{-11}. However, the rapid progress in the field of cooling and trapping of atoms and ions promises a major advance in high-resolution spectroscopy. Indeed, we have already witnessed a new visible standard based on an intercombination transition of cold Ca atoms.[5] An optical transition linewidth as narrow as a few tens of Hz (Q of 10^{13}) has also been observed using a cooled and trapped single $^{199}Hg+$ ion at 282 nm.[6] Such narrow transitions provided by free atoms or ions could offer an ultimate optical frequency standard free from virtually all of the conventional shifts and broadenings to the part in 10^{16} level.

While systems involved with laser cooling and trapping are expected to offer the highest quality optical frequency sources, compact and low cost systems are competitive practical choices even though their performance may be a decade worse. Here we present such a system, which involves a solid-state laser (Nd:YAG) stabilized either on an I_2 transition at 532 nm (after frequency doubling) or directly on an overtone transition of HCCD molecule at 1.064 μm. The stability of the system has already been demonstrated at the level of 10^{-14} at 800 s.

Stabilizing lasers on molecular overtone resonances is a particularly exciting prospect. These vibrational-overtone lines provide rich spectra of reference grids in the visible domain, while they maintain the same ~kHz linewidths as their fundamental counterparts, limited by molecular fluorescent decay at the vibration frequency. Molecular ro-vibrational lines are also less sensitive to external perturbations. However, owing to the extreme weakness of these otherwise attractive overtone transitions, they were not adopted as suitable frequency references in the visible until recently.[7,8] (For example, in our stabilization work of Nd:YAG laser, the employed HCCD overtone transition strength is only about one millionth of the I_2 line.) In this paper we present our recent development of a new cavity-enhanced frequency modulation spectroscopy which has enabled us to achieve record high detection sensitivities. The resulting excellent S/N for these weak but narrow overtone lines has opened the door for realizing thousands of high quality frequency references in the visible and near infrared.

The realization of the stable frequency sources in the optical range has resulted in parallel development of absolute and precise frequency measurement in the visible and near-infrared spectral region. A few frequency synthesis chains have been developed to phase-coherently connect the frequency of optical references to the cesium primary standard. (For example, see references [9] and [10].) These synthesis chains are complex systems and can only cover some discrete frequency marks in the optical spectrum. Difference frequency of many terahertz could still remain between a targeted frequency and a known reference. Over the years,

several approaches have been proposed and tested in searching for reliable and simpler solutions to make coherent transfer over large optical frequency gaps. Some recent popular schemes include: frequency interval bisections[11], optical-parametric oscillations (OPO)[12], optical comb generations[13,14], sum-and-difference in near infrared[15], and four-wave mixing in laser diodes.[16] All these techniques rely on the principle of difference-frequency synthesis, in contrast to the frequency harmonic generation method normally used in traditional frequency chains. To deal with relatively smaller frequency gaps on the order of a few terahertz, optical frequency comb generators certainly provide the most direct and simple approach. An electro-optic crystal is strongly excited by velocity-matched microwave fields to produce a significant phase modulation index for the light passing through the crystal. This electro-optic modulator is then placed inside a low-loss optical cavity, which resonates with the carrier frequency and all subsequently generated sidebands. This approach greatly enhances the modulation efficiency and the result is a rich spectrum of equally spaced lines spanning a few THz. In this article we will present the comb generator developed in our laboratory and discuss its use in our frequency measurement schemes.

NOISE-IMMUNE CAVITY-ENHANCED OPTICAL HETERODYNE MOLECULAR SPECTROSCOPY ("NICE-OHMS")

The P(5) line of the $(\nu_2 + 3\nu_3)$ overtone band of the $^{12}C_2HD$ molecule lies within the tuning range of the Nd:YAG laser at 1.064 μm. Its transition dipole moment is 0.069 millidebye (1 debye = 3.33564×10^{-30} C·m, a typical atomic transition dipole moment) and the linear absorption is calculated to be 8.7×10^{-7}/cm·torr. This weak transition strength makes it difficult to obtain the necessary level of nonlinear interaction between the light and the molecules to yield the sub-Doppler saturated absorption resonance. For metrology work, we usually use a low gas pressure (a few millitorr, 1 torr = 133 Pa) to avoid excessive pressure-related shifts and broadenings. When the pressure is sufficiently low, the average molecules will be in a free-flight regime, where they pass through the light field without suffering any collisions. Even in this case, the corresponding saturation power (the molecules see the light field as a π-pulse) is calculated to be 51 W. Under these circumstances, placing molecules inside a high-finesse cavity[17] is beneficial in several aspects. The cavity extends the effective cell length by a factor of (Finesse·$2/\pi$) while it builds up sufficient internal power for saturation. This helps to limit both the source and detection power to reasonable levels. The geometrical self-cleaning and mode-matching of the two counter-propagating waves inside the cavity are important for obtaining narrow and unshifted resonance lines.[18] In addition, the cavity itself provides a ready reference for short-term laser stabilization.

The use of the sharp cavity resonance, however, presents a challenge for a simple and yet effective modulation recovery scheme in signal detection. One direct method is to lock the laser on the cavity and dither the cavity resonance around the molecular resonance. Indeed, this technique is employed when we want to calibrate the size of the saturated absorption signal. However, the modulation frequency is by far too low to get away from the technical noise. Additionally, the narrow cavity resonance effectively converts any small laser frequency noise into amplitude noise, thus deteriorating the detection S/N. In our new modulation and detection scheme, we frequency modulate the input laser beam at exactly the splitting frequency of the cavity free-spectral-range (FSR). The cavity-transmitted light is subsequently detected and demodulated. The small frequency variations of the laser will still lead to some amplitude fluctuations and small optical phase shifts of the transmitted carrier, but they will also have exactly the same effects on the sidebands which are transmitted on adjacent or nearby cavity axial orders. Therefore, the transmitted light still accurately represents an FM spectral triplet,

with minimal AM conversion due to the relative laser/cavity frequency jitter. Thus, the detection noise level can approach the intrinsic AM noise of the laser at the modulation frequency. When an intracavity molecular resonance is present around one of the three cavity modes which accept the triplet light field, it will weaken and phase-shift the corresponding FM component to upset the FM balance and thereby create an RF heterodyne signal in the cavity transmission.

In short, the aforementioned modulation strategy allows the cavity to greatly enhance the molecular signal without any noise penalty. Therefore, the detection sensitivity is directly enhanced by the factor of (Finesse·$2/\pi$), compared with the conventional FM spectroscopy.[19] We refer to this modulation/detection scheme as noise-immune cavity-enhanced optical heterodyne molecular spectroscopy, i.e. "NICE-OHMS."

Fig. 1 shows the general experimental setup. Although the extreme accuracy of laser/cavity locking is no longer required for the detection of molecular signal, the laser linewidth relative to the cavity needs to be sufficiently narrowed so that its full power spectrum can be efficiently coupled into the cavity. Additionally, this laser/cavity locking loop serves as the short-term stabilizer for the laser frequency. The cavity discrimination signal is derived from the Pound-Drever-Hall RF sideband technique.[20] The electro-optic modulator 2 (EOM2) produces the 4 MHz FM sidebands which are detected in cavity reflection. The frequency servo action is carried out by the laser's internal piezo-electric transducer (PZT) and an external stabilizer[21], which uses just an acousto-optic modulator (AOM). The AOM provides sufficient servo bandwidth while the PZT corrects any slow deviations of the laser frequency. Fig. 2 illustrates the frequency noise spectral densities of the cavity error signal when the laser is unlocked, locked by the laser PZT only, and locked by the combined effort of the PZT and the AOM. The superior servo system with the PZT and AOM produces a frequency noise spectral density of only ~−34 dB Hz/$\sqrt{}$ Hz (20 mHz/$\sqrt{}$ Hz). This indicates the laser's linewidth relative to the cavity is a mere 1.3 mHz.[22] This locking performance can be further improved as the shot-noise-limited frequency noise spectral density is shown to be only 0.2 mHz/$\sqrt{}$ Hz.

It is important to have a precision scanning capability for our spectrometer to perform high resolution studies of the signal lineshape, linewidth, and the line center. This is accomplished by the frequency-offset-locking loop shown in Fig. 1. The stable reference is provided by a second Nd:YAG laser which is frequency doubled and locked on an I_2 transition at 532 nm.[23] The heterodyne beat signal between the two lasers is phase-locked to a synthesizer. To improve S/N and baseline stability, we modulate the cavity frequency around the molecular resonance by an FM dither on the synthesizer and use an audio lock-in for signal recovery. The same synthesizer is then frequency stepped by a computer to force the cavity/laser system to scan out the molecular resonance. Signal averaging over many scans (~one minute each) is available with the long-term stability of the laser/I_2 frequency reference system in the sub-10-Hz domain.

Phase modulation of the laser beam at the FSR frequency is generated by the resonantly coupled EOM1 and detected in cavity transmission by an InGaAs p-i-n diode in a resonant RF tank circuit. The modulation signal is provided by a low-phase-noise crystal voltage-controlled-oscillator (VCO). The modulation index of ~0.5 is chosen so that the central carrier still effectively saturates the molecular absorption, while the sidebands are large enough to give a strong heterodyne cross-term. An isolation AOM directly after the cavity prevents optical feedback from the detector. Subsequent phase sensitive RF demodulation yields the molecular dispersion signal. During the cavity scan, its FSR will change slightly. To maintain the noise-immune property, we actively track the VCO frequency to the cavity FSR by applying a small frequency dither on the VCO, which is then also detected in cavity reflection. This control loop is also shown in Fig. 1.

Synthesized HCCD gas (81%/19% HCCD/HCCH by integration of the ^{1}H NMR spectrum, ~55% chemical purity by a residual gas analyzer) is placed inside a cavity with an FSR of 319.695 MHz and finesse of 31,900. One mirror is flat, the other has a 1 m radius of curvature. The intracavity beam waist is ~0.410 mm, dictating the room temperature transit time limit of 270 kHz FWHM.[24] For our 10 mtorr sample gas, the linear absorption through our 46.9 cm long cavity (single pass) is 1.6×10^{-7}, leading to an absorption coefficient of 6.2×10^{-7}/torr·cm for pure HCCD, in good agreement with the available band strength data.[25] The cavity has a resonant transmission efficiency of 41%, which decreases by 1.3×10^{-3} near the Doppler profile peak (a contrast of 0.32%). Tuning onto the saturation resonance increases the maximum transmission by 138×10^{-6}, corresponding to a saturation depth of 10.6%. From a 75 mW input light, the cavity has a power buildup to 300 W, giving a saturation parameter of ~0.46.

In our frequency stabilization experiment, we usually use the central carrier to interact with the molecules. This central component is intrinsically sensitive only to the dispersion part of the molecular resonance, independent of the detection phase of the RF local oscillator.[19] The additional dither modulation on the cavity resonance itself requires an audio lock-in to further process the RF-demodulated signal, and the resultant signal lineshape shows a derivative form of dispersion. Fig. 3 shows a typical scan of the overtone resonance with 5.3 mtorr sample gas and a 640 kHz peak-to-peak cavity dither. The lineshape model is based on Wahlquist's modulation-broadening formalism for a dispersion signal.[26] The diminutive fit residual, after a 10 times magnification, indicates a well-understood lineshape in our spectrometer. The transition linewidth of 705 kHz (after removal of the modulation broadening by the fit) is due to the power- and pressure-broadenings of the 270 kHz transit time linewidth. The power-broadening contribution to the linewidth can be removed by knowing the saturation level at the operating pressure. The pressure broadening (FWHM) rate is then determined to be 34.7±0.8 MHz/torr. The zero-pressure, zero-intensity extrapolated linewidth is 290±7 kHz, very near the value of 270 kHz set by the transit time. We can summarize these results by an expression for the saturation intensity: $I_{sat} = 6103\ (\Gamma_T + 34.7\ P)^2$ W/mm^2, where Γ_T is the FWHM (in MHz) associated with the transit time, and P is the pressure in torr. The equivalent intracavity saturation power in the free-flight regime is 117 W, reduced to 29.3 mW input by the cavity enhancement of 4,000. We have also measured a pressure shift of the line center ~+250 (± 20%) Hz/mtorr with our current sample gas.

To obtain a proper discrimination lineshape of the molecular resonance for the laser to lock onto, we use the second harmonic detection on the audio lock-in to obtain a second derivative of dispersion. This is shown in Fig. 4 with 10 mtorr gas. With the saturated absorption in the 1.7×10^{-8} domain, we obtained a S/N of 8,700 at 1 s averaging, ~2 times above the calculated shot noise limit. This corresponds to a noise-equivalent detection sensitivity of 2×10^{-12} for integrated absorption at 1-s averaging. (Recently we have further improved this sensitivity to 6×10^{-13} by using an improved cavity.) This level of S/N sets the frequency rms noise when the laser is locked on the molecular line.

FREQUENCY STABILIZATION RESULTS

For testing the quality of this C_2HD overtone resonance as an optical frequency standard at 1.064 μm, we lock the laser/cavity system onto the line and measure the heterodyne beat against the known Nd:YAG/I2 reference system.[23] The molecular error signal is integrated and then fed onto the cavity PZT to maintain the proper cavity length. We find 10 mtorr is the optimum gas pressure for a maximum slope of the molecular locking error signal, by taking into account the combined parameters of linear absorption, saturated absorption hole

depth and the pressure-broadened linewidth. In Fig. 5 the counted beat frequency vs. time shows a drift ~20 Hz/h. The second Nd:YAG laser (after frequency doubling) is locked on I_2: R(56) 32-0, component a_{10}.[23] With the mean value of the beat frequency between the two lasers at 5252.2261±0.0026 MHz, we determine the absolute frequency of the P(5) line in the $(\nu_2+3\nu_3)$ band of the $^{12}C_2HD$ to be 281,635,363.962 MHz±20.2 kHz. The 20 kHz uncertainty is mainly due to the limited knowledge of the absolute frequency of the Nd:YAG/I_2, a secondary standard. At 1-s averaging we obtain a frequency noise of ±100 Hz, in direct agreement with the S/N available at 1.064 μm. (The Nd:YAG/I_2 reference system has a 20 Hz rms, 8×10^{-14} frequency noise at 1 second as tested with a second I_2 spectrometer.) The corresponding Allan variance of $\sigma_y = 3.4\times10^{-13}/\sqrt{\tau}$ improves to 1×10^{-14} at a longer integration time (> 800 s), a promising indicator for an ultrastable frequency reference. The visible noise bump around ~200 s on the Allan variance is associated with the lab room temperature cycling period.

Linewidth narrowing is offered by slow molecules since the natural lifetime of the overtone transition is about 300 times longer than our current transit time.[27] The cavity input power is reduced 75 times from our maximum available power to 1 mW. A low power is necessary so that the low Rabi frequency leads to appreciable saturation only for the slowest molecules. In our < 2 mtorr sample gas, the mean-free-path of molecules is ~30 times longer than the transverse field dimension, thereby creating the so-called transit-time regime. The saturation now becomes inhomogeneous, with molecules from different transverse velocity groups contributing different intensities and widths. Slow molecules which spend their whole lifetime inside the field will have a constant and velocity-independent saturation parameter, controlled primarily by the collisional broadening. Considering their shorter interaction times, faster moving molecules will see a reduced saturation and will mostly contribute to the wings of the resonance. Fig. 6 shows a resonance with a linewidth of ~20 kHz, without correction for the modulation broadening by a 30 kHz peak-to-peak dither of the cavity. This is 13 times narrower than that set by the room temperature transit-time-limit, and is mainly limited by the relatively high pressure (1.8 mtorr). At present the limited S/N associated with the low power has prevented us from taking full advantage of this narrow linewidth. With an improved system this approach will enable us to access the information of free molecules with minimized second order Doppler shift (~2×10^{-14}), thereby creating an optical frequency standard of potentially high accuracy.

OPTICAL FREQUENCY COMB GENERATORS

An optical frequency comb (OFC) generator is a simple system employing only one laser. Yet it offers a unique property of supplying a comb of equally spaced spectral lines around the carrier. These lines are modulation sidebands generated by an electro-optic modulator (EOM). To enhance the optical-RF field interactions, the EOM is placed inside a low loss optical cavity in resonance with the carrier and all the sidebands. In other words, the RF modulation frequency equals an integer multiple of the cavity FSR. In principle, the span of the generated comb is limited only by the system dispersion, which can be carefully compensated following designs in ultra-fast laser systems. A 4-THz wide OFC has already been observed at 1.5 μm[28], showing the possibility of shifting 2% of the optical frequency in a single step. We note that an appropriately low-noise RF oscillator should be used to drive the EOM so that high-order sidebands do not quickly collapse due to the multiplied phase noise amplitude.

The power spectrum of the OFC is shown[28] to be proportional to an exponential function. Denoting P_k as the power of the kth sideband, we have $P_k \propto exp(-|k|\pi/\beta F)$ where β is the modulation index of the EOM and F is the finesse of the crystal-loaded cavity. To improve the efficiency of the comb generator and to have a single pure spectral line output for optical

frequency metrology, we replace the cavity output mirror with a short filter cavity to resonantly output an individual sideband from the comb. If the FSR of this filter cavity is larger than the comb width, then the filter will be resonance-free until one reaches the desired sideband. Therefore, the filter cavity will not alter the comb generation process until a good match occurs between its resonance and a sideband, beyond which the comb spectrum will be sharply cut off. The filtered single spectral line can be conveniently detected by heterodyne-mixing with a tunable laser source. Since we extract the full power of the chosen sideband out of the comb generator while keeping the carrier and all other sidebands trapped inside, we can expect an important improvement of the detection S/N. Using a filter cavity not only resonantly increases the signal size of the desired sideband output, but also reduces the detected noise level as the larger DC powers distributed among the carrier and lower order sidebands are not detected.

In this experiment we use a prototype EOM.[29] It consists of a broadband antireflection coated Mg:LiNbO3 crystal (2x1x35.4-mm) embedded in a resonant microwave cavity. The cavity design employs a waveguide geometry to force the match between the microwave phase velocity and the optical group velocity through the crystal. The microwave resonance at 10.5 GHz has a bandwidth of ~0.3 GHz and a Q factor of 230. A modulation index of ~0.8 is obtained with a microwave power of 0.6 W. This EOM is placed inside our three-mirror cavity, as shown in Fig. 7. All three mirrors are identical lens substrates with an effective focal length of 25 cm. The convex faces are antireflection coated at 633 nm, while the flat faces are coated to have high reflectivity, about 99.6%. With two such mirrors (M1 and M2) we build a cavity with a finesse of 680 and a transmission efficiency of 20%, implying a transmission coefficient (T) of 0.2% for each mirror. The cavity FSR is 1/16 of the EOM RF frequency. When loaded with the cold crystal, the finesse and efficiency drop to 200 and 2%, respectively, corresponding to a 1.1% one-way loss through the modulator. Turning on the RF power to the EOM further drops the cavity efficiency to 0.15% for the overall modulated output, due to the increased mismatch of input coupling when sideband generation enhances the carrier loss. The filter cavity formed by mirrors M2 and M3 has a finesse of 400, a FSR of ~2 THz, and an efficiency ~30%, and increases the output power of the selected sideband by a factor of 150. The PZT mounted on the filter cavity output mirror M3 is used to tune the filter bandpass frequency. Approximately 150 μW power of a polarization-stabilized He-Ne laser is incident on the comb generator. Part of the output light from the OFC generator is monitored by a DC photo detector, while the other part is sent to an avalanche photodiode (APD) for heterodyne-mixing with an external-cavity tunable diode laser at 633 nm.

Fig. 8 shows the DC-monitored output spectrum of our OFC generator as we continuously tune the filter cavity resonance over part of the comb spectrum. A comb span wider than 1 THz is clearly visible from one side of the carrier frequency. The filter cavity has a FWHM of ~5 GHz. This gave enough resolution to resolve individual sidebands spaced 10.5 GHz apart. Based on the observation that high-order (~100th) sidebands still have a good S/N, we expect to see a wider comb with a filter cavity having a larger FSR. (It will also need a higher finesse to maintain its resolution.) The slope on this comb spectrum is roughly 16 dB/THz.

About 15 μW power from an external cavity tunable 633 nm diode laser is used for the heterodyne detection of the OFC sideband. Fig. 9 shows the resulting beat spectrum. The filter cavity resonance is subsequently tuned onto the 48th (505 GHz), 96th (1.01 THz), and 144th (1.515 THz) sideband of the He-Ne laser. In a 100 kHz bandwidth we obtain S/N of 35 dB, 26 dB, and 20 dB, respectively. The noise floor is fixed by the shot noise of the detected light power, increased by the APD's excessive noise factor. These beat signals can be easily counted via a tracking-filter comprised of a voltage-controlled RF oscillator phase-locked onto the beat signal.

As the filter cavity selects out a particular sideband, it causes little effect on the lower order sidebands being generated inside the comb generator. However, once the energy in a sideband is coupled out, the comb generation beyond that is strongly reduced. This mechanism is confirmed in the following way. We park the filter cavity resonance on top of the 48th sideband, and then we position the diode laser frequency successively to be in line with the 47th, 48th, and 49th sideband. Heterodyne detection shows that a fraction of the 47th sideband power leaked out due to the finite width of the pass-filter (-17.7 dB below the 48th sideband). The magnitude of the 49th sideband is lower by 5.6 dB, -23.3 dB relative to the 48th sideband. This good spectral purity will improve further using a filter cavity of higher efficiency or better finesse.

CONCLUSIONS

The experimental demonstration of the noise-immune cavity-enhanced optical heterodyne molecular spectroscopy (NICE-OHMS) has firmly established itself among the most sensitive detection methods currently available. Its powerful utilities in the laser frequency stabilization and ultra-high resolution spectroscopy are also indispensable for realizing thousands of molecular overtone transitions in the visible and near-IR as high-quality optical frequency/wavelength references. The work of Nd:YAG laser frequency stabilization using the P(5) transition in the C_2HD ($\nu_2+3\nu_3$) overtone band at 1.064 μm has achieved similar results to that using iodine transitions with modulation transfer spectroscopy,[23] despite the fact that the C_2HD transition is a million times weaker than that of iodine. Slow molecules optically selected should provide more accurate information about the resonance line center of free molecules. The relative stability of this system is already better than most visible frequency standards, including those recommended by CIPM/CCDM in 1992.[4] Potential improvements on our system would be to use larger mode-size and higher finesse cavities to further enhance the resolution and sensitivity.

We have realized a wide span (> 3 THz) optical frequency comb generator. We improve the comb generator efficiency by replacing the output mirror with a short filter cavity to allow efficient escape of the selected comb component. With limited power available from a He-Ne laser, we are able to demonstrate a 1.5 terahertz heterodyne beat signal with a S/N of 20 dB at 100 kHz bandwidth. We intend to use this OFC generator to bridge gaps between stronger and spectrally narrower iodine molecule absorption lines around 633 nm and the R(127) transition where the He-Ne laser is traditionally stabilized. An interesting Neon transition (1S5 -> 2P8) at 633.6 nm can also be measured in its absolute frequency. We are also planning to improve our frequency chain for the green iodine transitions at 532 nm.[23] We are in the process of establishing grids of molecular ro-vibrational lines as frequency references over the red part of the visible spectrum. As the spacing between adjacent rotational lines usually lies anywhere between a few hundred GHz to a few THz, this OFC generator covering THz frequency gaps becomes an essential part of our phase coherent frequency chains.

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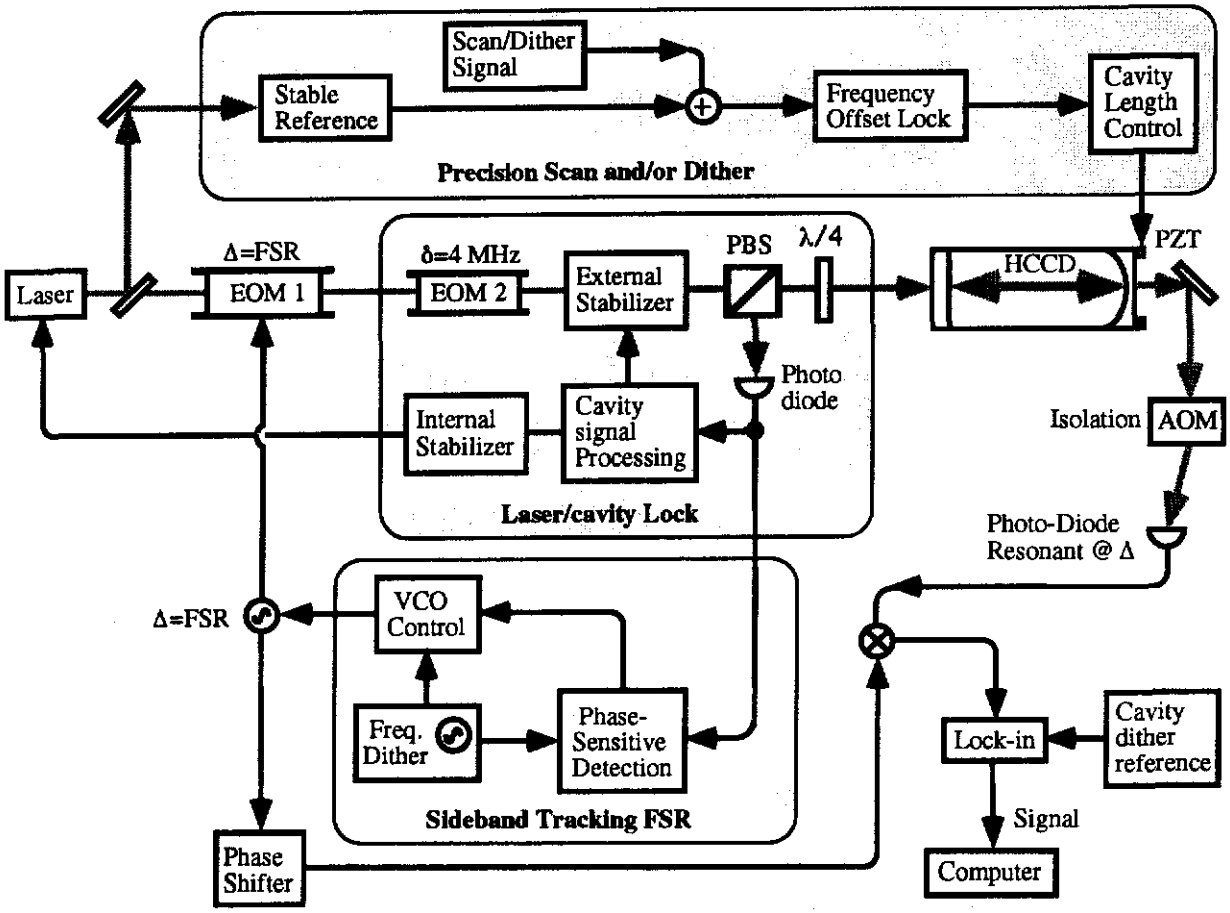

Figure 1 General experimental setup for the NICE-OHMS spectrometer.

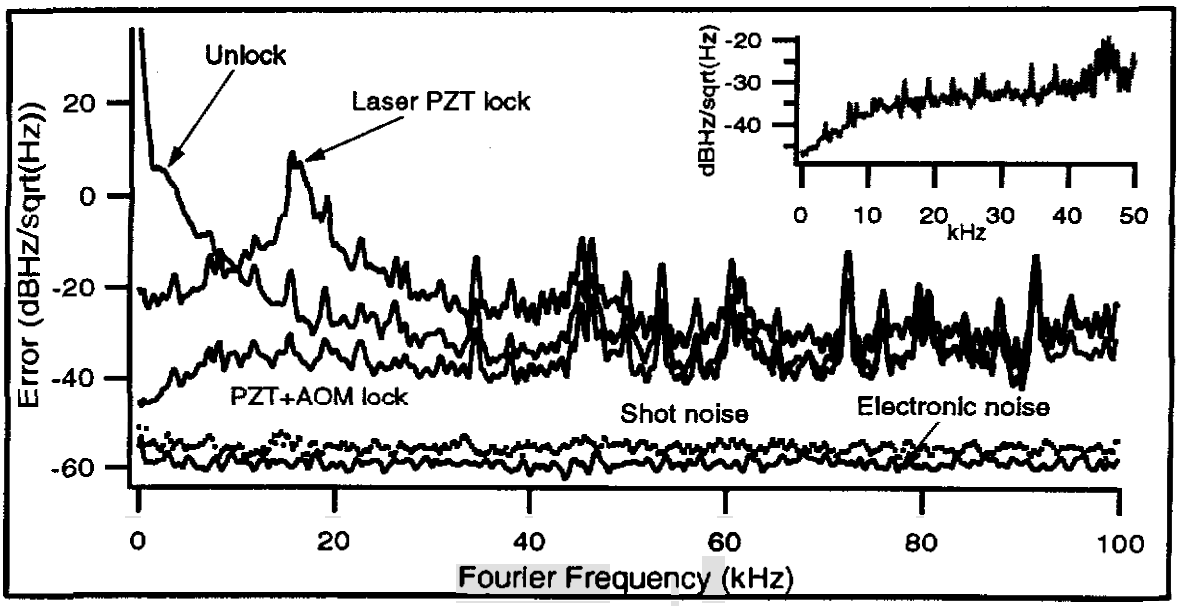

Figure 2 Laser/cavity locking frequency noise spectral density.

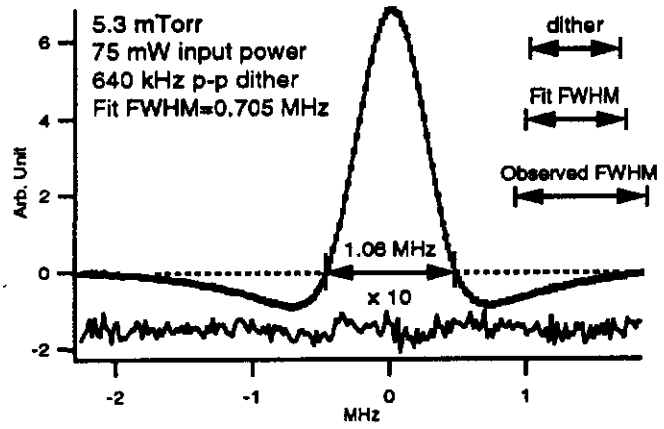

Figure 3 Frequency scan of the C_2HD ($\nu_2 + 3\nu_3$) P(5) line and overlaid theoretical fit, with fit residuals magnified by 10 times.

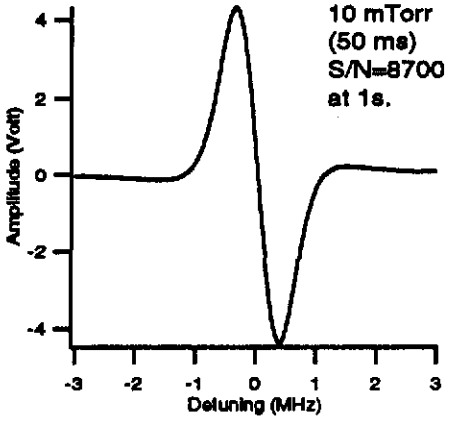

Figure 4 Molecular discrimination signal used for stabilizing Nd:YAG laser, obtained by the 2nd harmonic detection of the molecular dispersion.

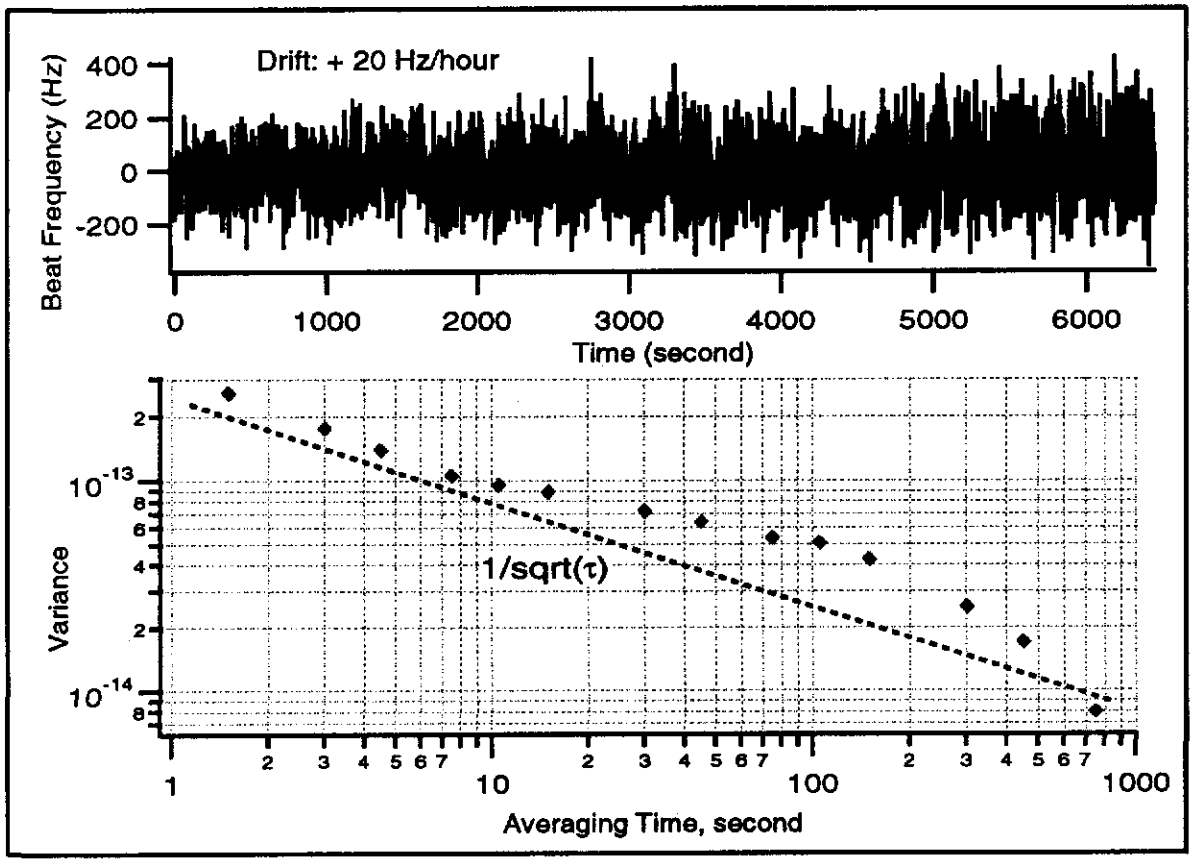

Figure 5 Time record of the beat frequency between two stabilized lasers and the Allan variance.

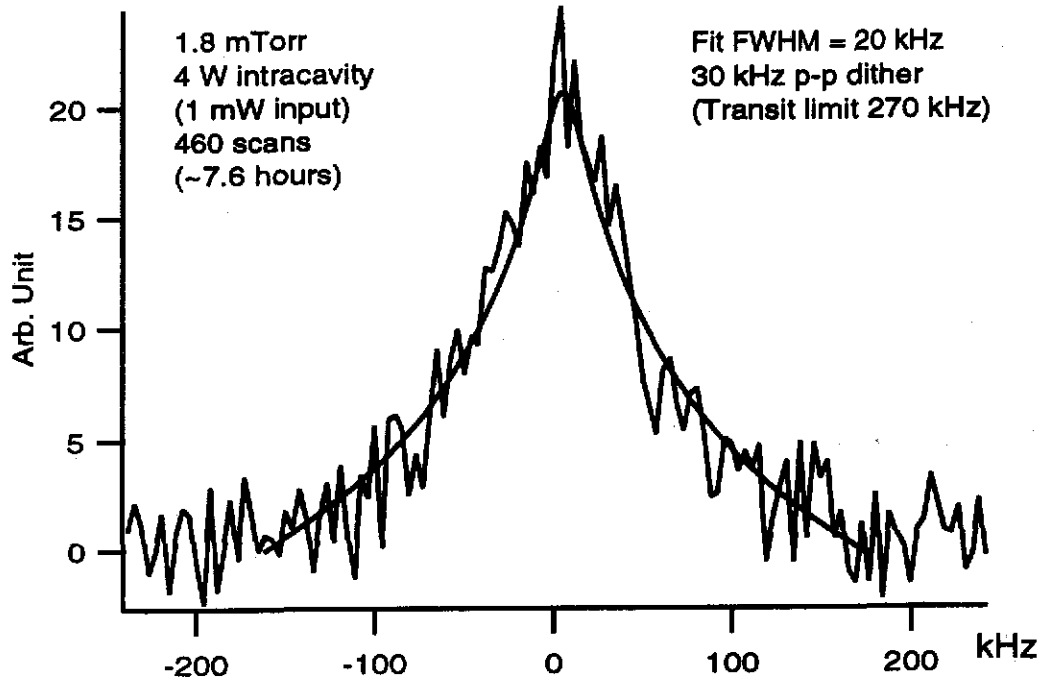

Figure 6 With low power and gas pressure, slow molecules give a linewidth 13 times below the transit limit.

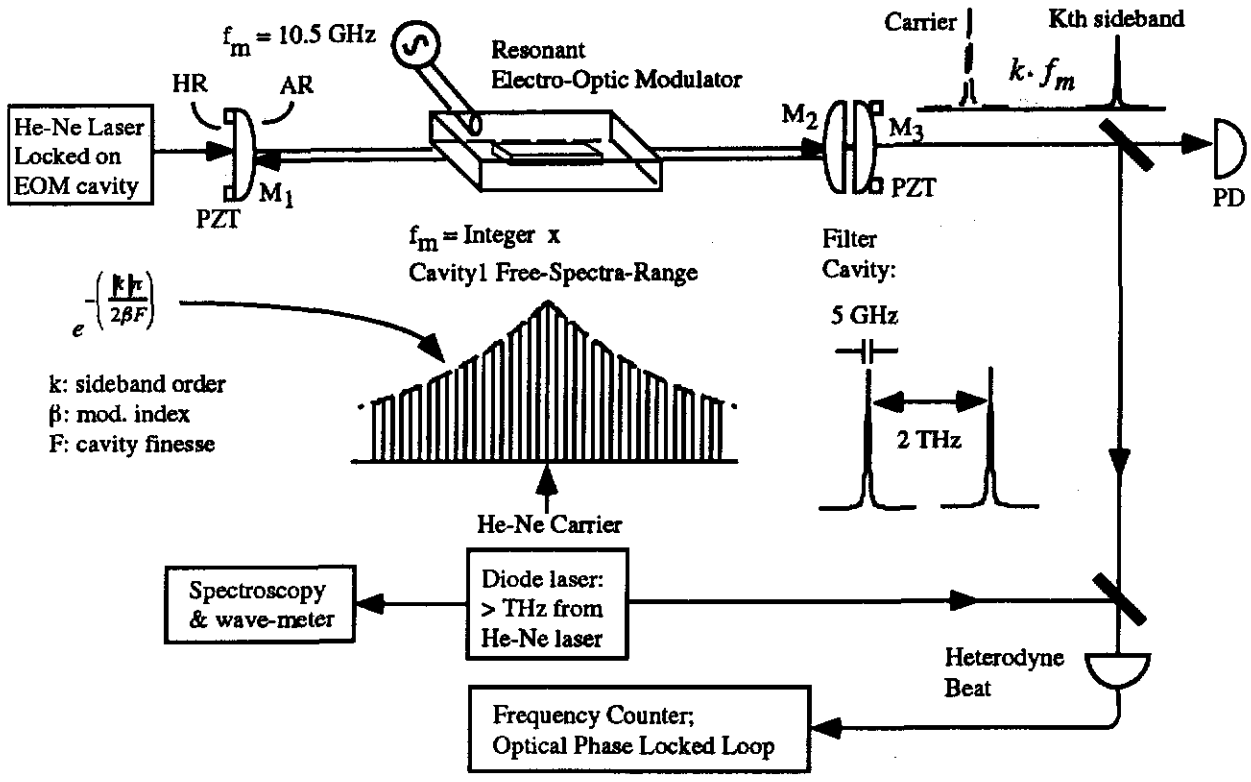

Figure 7 Schematics for optical frequency comb generator.

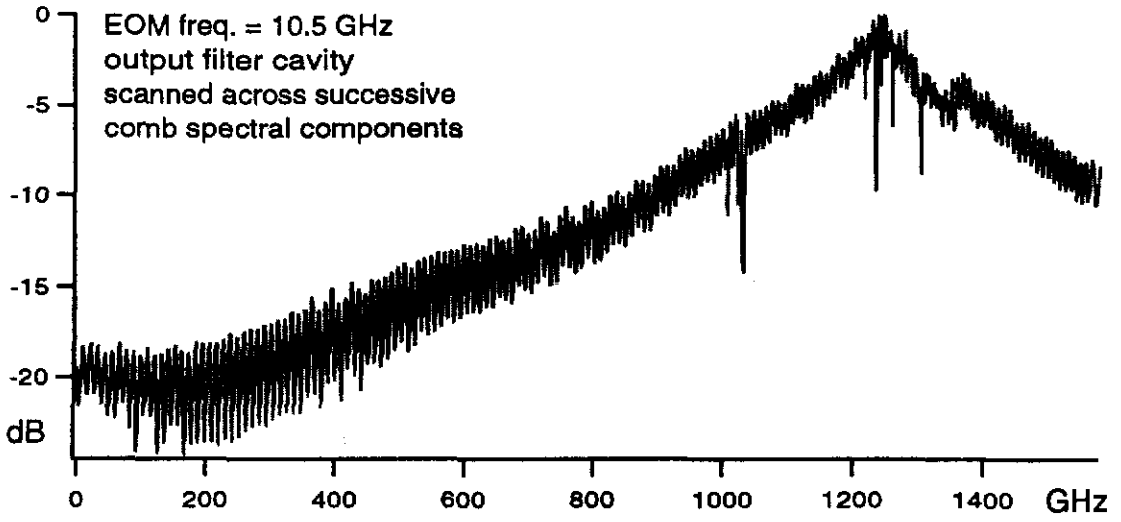

Figure 8 Optical comb generator output spectrum as the filter cavity resonance is scanned through the comb spectrum. Comb line spacing is 10.5 GHz.

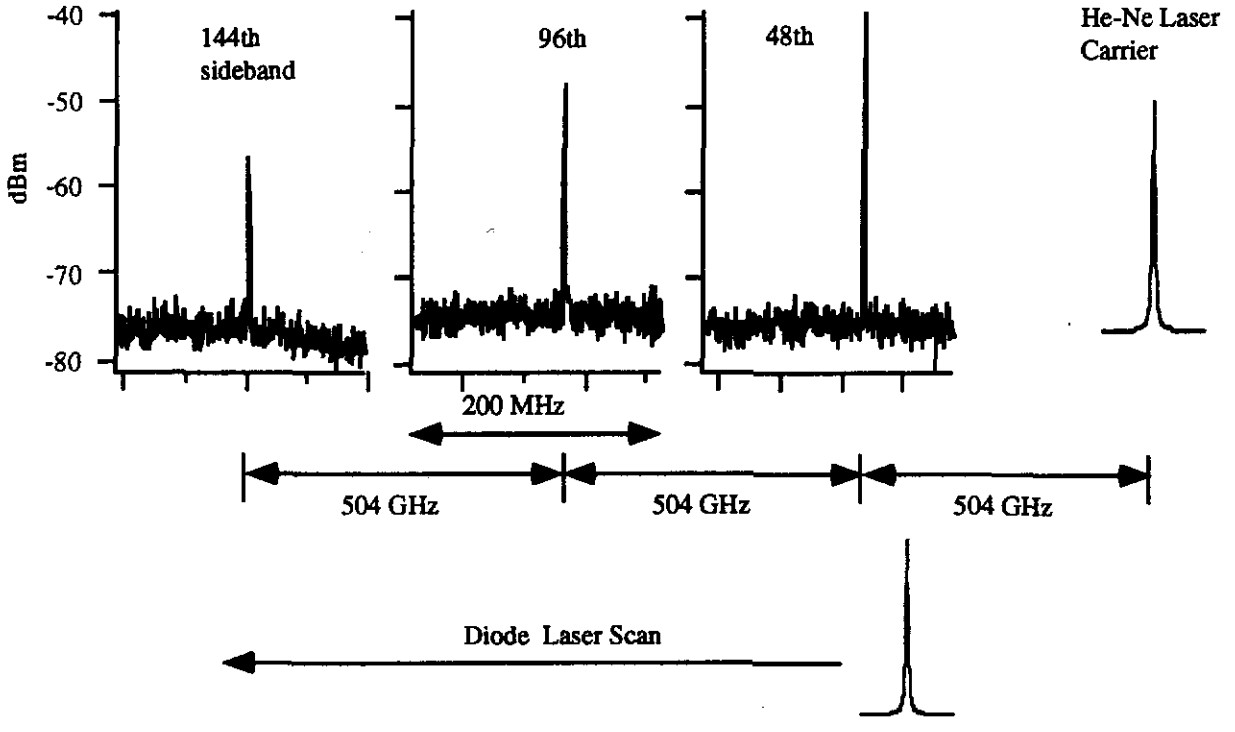

Figure 9 Beat between the diode laser and the 48th (505 GHz), 96th (1.01 THz), and 144th (1.515 THz) sidebands of the He-Ne laser. All with 100 kHz resolution bandwidth.

Questions and Answers

DEMETRIOS MATSAKIS (USNO): I notice some deviations on your Allan plot at about 100 seconds. Do you know what caused them?

JUN YE: Demetrios is talking about this little bump on the Allan variance. And if you really try to make a line, this line actually follows very nicely to 1 over the square root of τ. That means it's a white noise. However, there's a bump that shows up. That means you have a sinusoidal modulation on your beep frequency. And really if you use a digital low-pass filter (such as that by the decimation process) to process the time record of the beat frequency, the small sinusoidal modulation will emerge from the noise.

The problem with this 5-minute kind of modulation is due to the air temperature cycling our laboratory. When the temperature changes in the lab, it will affect the optical path lengths of some residual interferometers within our spectrometer. For example, the thickness of a glass window could change. The change of these residual fringes could shift the stabilized optical frequency. And that shows up as a modulation in our beat frequency record.